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Disclaimer

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The ^{242m}Am isomer, a well-known candidate for photo-depopulation research, has been studied in this first ever Coulomb excitation of a nearly pure ($\approx 98\%$) isomer target. Thirty new states, including a new rotational band built on a $K^\pi = 6^-$ state have been identified. Strong K-mixing results in nearly equal populations of the $K^\pi = 5^-$ and 6^- states. Newly identified states have been assigned to the $K^\pi = 3^-$ rotational band, the lowest states of which are known to decay into the ground-state band. Implications regarding K-mixing and Coulomb excitation paths to the ground state are discussed.

I. INTRODUCTION

The ^{242}Am nucleus has a 48.6 keV, $K^\pi = 5^-$ isomer (^{242m}Am, $t_{1/2} = 141$ y) which is known to decay by a 99.55% electromagnetic (EM) decay branch to the ground state [1, 2] (Figure 1). This isomer has drawn considerable interest related to the possibility of energy storage and controlled release [3–5] by absorption of an x-ray photon. Theoretical estimates of small de-excitation γ-ray yields have been made, based on the scenario of 4 keV E2 excitation by real photons to the 53 keV $I^\pi K = 3^- 0$ state of the ground-state band (GSB) [6], although this transition has not been observed directly. The successful photo-depopulation of the 75 keV ^{180m}Ta isomer [7] has increased attention to the subject.

Nuclei in the $A \approx 240$ mass region are known to have prolate shapes [8]. For these axially symmetric deformed nuclei, K, the projection of the total spin I on the symmetry axis, is expected to be a good quantum number. The K-selection rule [8] forbids EM transitions between two states $| I_i M_i K_i \rangle$ and $| I_f M_f K_f \rangle$ for which the forbiddenness $\nu \equiv | \Delta K | - \lambda$ is greater than zero, where λ is the multipole order and $\Delta K \equiv K_f - K_i$. The degree of hindrance of a K-forbidden transition can be expressed in terms of the "reduced hindrance,"

$$f_\nu \equiv (B(\mathcal{M}\lambda)_{\mathrm{W.u.}}/B(\mathcal{M}\lambda))^{\frac{1}{\nu}}, \qquad (1)$$

where $B(\mathcal{M}\lambda)_{\mathrm{W.u.}}$ is the Weisskopf single-particle estimate of the EM reduced transition probability $B(\mathcal{M}\lambda)$.
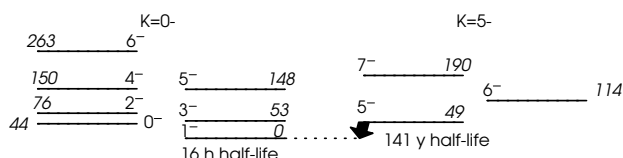

FIG. 1: Low-lying states in ^{242}Am [2] and their energies in keV showing the known EM decay of the 5^- isomer [1].

For K-forbidden transitions, f_ν is expected to be $\gg 1$, and this is the case for the $\nu = 1$ E4 isomer decay, which has $f_\nu \sim 10^5$. Recent work on the prolate quadrupole-deformed ^{178}Hf nucleus has demonstrated that K-mixing increases rapidly with spin I in the low-K bands, whereas the high-K bands were found to remain very pure in K [9]. This spin-dependent mixing made Coulomb excitation of rotational bands built on high-K $6^+, 8^-$ and 16^+ isomers of ^{178}Hf possible. The study of EM excitation and de-excitation of high-K isomeric states has demonstrated significant violations of the K-selection rule in a number of axially symmetric, quadrupole-deformed nuclei [7, 9–13] and suggested a number of different mechanisms of K-violation [10, 11, 14–20].

The present experiment was devised to search for states coupled to the ^{242m}Am isomer, in particular those which might provide paths to the ground state, and to measure the EM transition matrix elements coupling the isomer to other states. Compared to excitation of the isomer from the ground state, the nearly pure isomer target provides a significant increase in sensitivity to the relevant matrix elements and, consequently, an opportunity to understand K-mixing in the $A = 242$ region.

II. EXPERIMENT

A $\approx 98\%$ enriched ^{242m}Am isomer sample was separated from contaminants and decay products, and electrodeposited onto a 5 mg/cm^2 natural Ni foil at Lawrence Livermore National Laboratory (U.S.A.). The 2% contaminant was predominantly ^{241}Am. This target was Coulomb excited by a 170.5 MeV ^{40}Ar beam using the ATLAS Linac at Argonne National Laboratory (U.S.A.). The de-excitation γ-rays were detected by Gammasphere, a 4π 110-element Compton-suppressed high-purity germanium (HPGe) array, of which 101 detectors were installed. In addition, five low-energy photon spectrometers (LEPS) were installed in Gammasphere to provide
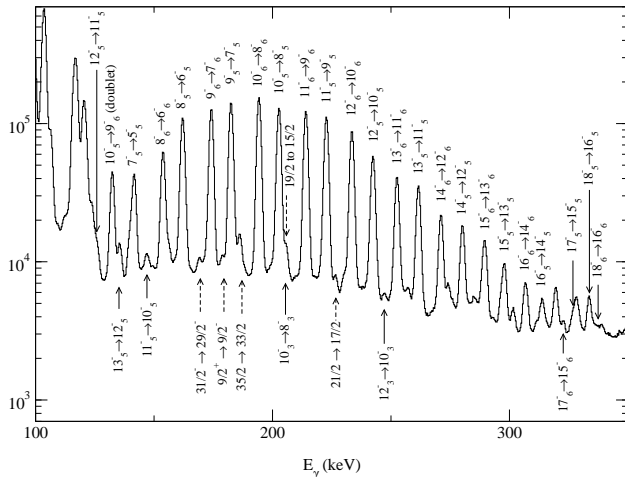
FIG. 2: A partial unsubtracted γ-ray energy spectrum of particle-γ singles with known transitions labeled with the initial and final I_K^π values. Transitions known to belong to ^{241}Am are indicated by the dashed arrows. The unlabeled low-energy peaks are dominated by x-rays from the target activity.

higher efficiency for transitions with $E \lesssim 300$ keV and sufficient resolution to identify two K X-ray transitions in Am atoms excited by the beam. The 500 μg/cm^2 target had a calculated activity of ≈ 1.6 milliCurie. In order to reduce the extremely high rate of random γ-ray coincidences, events were triggered by a coincidence between a backscattered Ar ion detected by CHICO [21], Rochester's parallel plate avalanche counter (PPAC), and at least one clean photon in Gammasphere or a LEPS. The combined power of CHICO plus Gammasphere produced clean spectra (Figure 2) despite the total count rates of ~ 500 kHz from x-rays in Gammasphere and ~ 1 MHz from α decays in CHICO due to the natural activity of the target.

During 106 hours of beam at ~ 0.3 particle nano-Amp and charge state 9^+, approximately 3×10^7 particle, γ-ray (p-γ) events were recorded. Doppler-shift correction of the de-excitation γ rays was not necessary, since the recoiling ^{242}Am ions were stopped in the target's Ni backing prior to γ decay. A single-γ spectrum (Figure 2) and a γ-γ matrix were constructed from the p-γ events and used to build a level scheme (Figure 3) by gating on one γ-ray peak and observing coincident γ-ray energies. There were an insufficient number of γ-ray events to construct useful higher-dimensional data sets.

III. ANALYSIS

The single-γ spectrum, which closely resembles that of a single rotational band (Figure 2), was determined to be the result of two rotational bands, populated with remarkably similar strengths. The first band, built on the $K^\pi = 5^-$ isomer at 48.6 keV and previously known up

to spin 7^-, was extended to a tentative $I^\pi = 18^-$ state (Figure 3). The second band is identified as a $K^\pi = 6^-$ band by its spin values, which were determined by γ-γ coincidences with transitions in the 5^- band due to highly-converted γ-decay feeding. Coincidences between γ-rays in the 5^- and 6^- bands were used to measure the internal conversion decay branches between the two bands. Upper and lower limits on the energies of the $K^\pi = 6^-$ levels were deduced by comparing discontinuities in the conversion branches to the known energies of the K- and L-edges in the internal conversion coefficients [23]. Two highly-converted interband γ-ray transitions and possibly a third with the correct energy difference were observed feeding the $K^\pi = 6^-$ band from the $K^\pi = 5^-$ band and used to set the energy of the 6^- state at 100.4(7) keV, consistent with the upper and lower limits. This makes the $K^\pi = 6^-$ band *yrast* with the $I^\pi K = 6^-6$ state 14 keV below the $I^\pi K = 6^-5$ state. The present 6^- band head energy is further supported by the previous observation of two unassigned states at 99 and 171 keV using a ^{243}Am(d,t)^{242}Am reaction [24]. These are now believed to be the 6^- and 7^- states of the $K = 6$ band (Figure 3). A previous calculation predicted an energy of 126(20) keV for the $I^\pi K = 6^-6$ state [22]. Higher-spin levels ($8^- \leq I^\pi \leq 14^-$ and a tentative 15^- level) of the $K^\pi = 3^-$ rotational band were identified by comparing the energies and moments of inertia to those of the known $K^\pi = 3^-$ levels (Figure 4) and all other previously known states of appropriate spin and energy. The tentative spin assignments were deduced from the pattern of stretched E2 transitions to the $K^\pi = 5^-$ band.

A relative photopeak efficiency curve was obtained from ^{152}Eu, ^{182}Ta, ^{243}Am and ^{56}Co source data. It was determined that the high activity of the target caused considerable dead time in Gammasphere, lowering the effective *absolute* photopeak efficiency of the array from the expected value of 8.9(2)% [25] to 5.6(4)% at 1.333 MeV. This absolute efficiency was obtained by measuring the ratio of mutual Coulomb excitation of the ^{40}Ar beam and the ^{242}Am target to the total population of the ^{242}Am states. The ^{40}Ar 2_1^+ Coulomb excitation cross section was calculated using the known $B(E2)$ values [26] and compared to the measured value, arriving at the absolute efficiency 5.2(4)% at 1.461 MeV. The relative efficiency curve was normalized to this value.

Relative intensities of the strongest γ-decay transitions were measured directly from the γ-ray singles and the γ-γ matrix. The branching ratios of the $K^\pi = 5^-, 6^-$ states were obtained by setting gates on γ-ray transitions above the states and correcting the resultant yields below each branch for the absolute photopeak efficiency and gate effects. The branching ratios were measured for unobserved γ decays with a sensitivity of $\approx 5\%$ of the total decay width of each state. The ratio of the $\Delta I = 1$ to $\Delta I = 2$ intraband transition intensities were used to determine $|g_K - g_R|/Q_0$ values for the $K^\pi = 5^-$ and 6^- bands. Assuming that the Alaga rule is valid for the intraband transitions, i.e., that the mixing of states can be
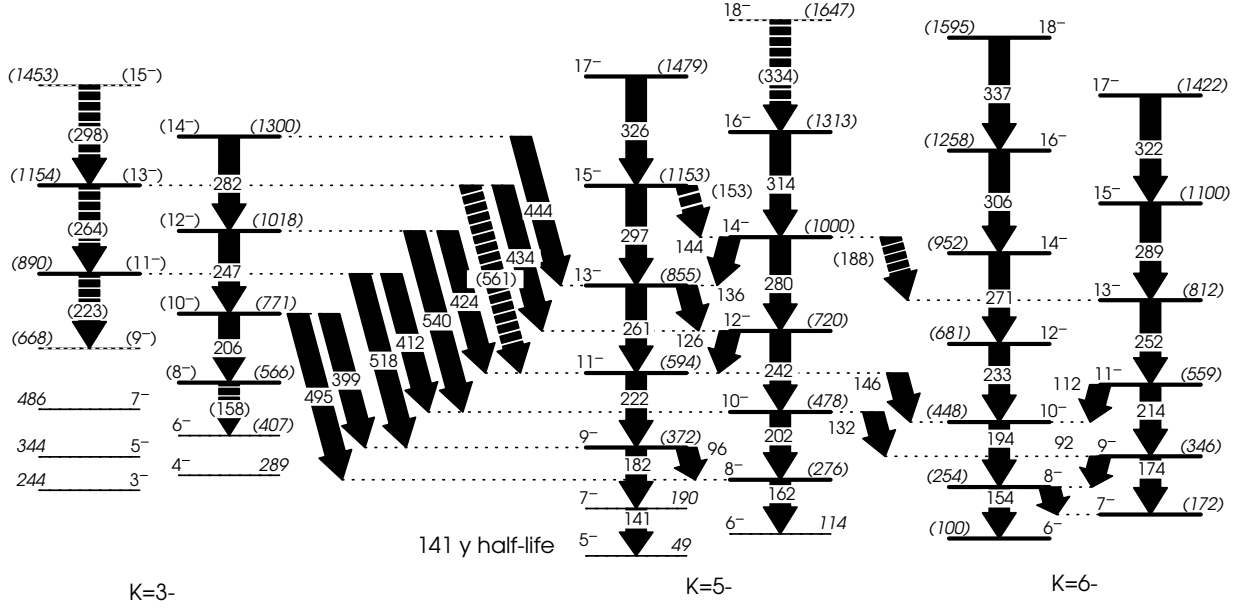
FIG. 3: A partial level scheme for ^{242}Am. Previously known states (thin lines) are from [22]. Energies of new levels from the present work (bold lines) have errors of ≤ 2 keV.
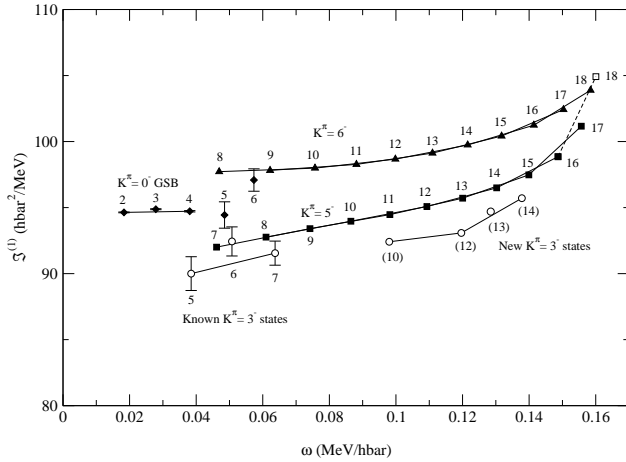

FIG. 4: Moments of inertia for the known $K^{\pi} = 0^{-}, 3^{-}$ ([22, 24]), $5^{-}, 6^{-}$ (present work) bands and the newly assigned $K^{\pi} = 3^{-}$ states. Errors are approximately the size of the points, except where shown by bars.

FIG. 5: Measured (points) and calculated (lines) intraband γ-ray yields in the $K^{\pi} = 5^{-}$ $K^{\pi} = 6^{-}$ bands from the model-dependent fit of matrix elements described in the text.

ignored in the angular momentum coupling, it was found that $\mid g_{K} - g_{R} \mid /Q_{0} \approx 0.014, 0.004$ for the $K^{\pi} = 5^{-}, 6^{-}$ bands respectively. The coupled-channel, semiclassical Coulomb excitation search code GOSIA [27] was used to fit intrinsic E2 matrix elements to the measured γ-ray yield data by χ^2 minimization. In this first iteration, the matrix elements coupling the $K^{\pi} = 5^{-}, 6^{-}$ and 3^{-} bands were fit to the measured γ-ray yields (Figures 5, 6), assuming the Alaga rule for interband transitions (i.e., neglecting band mixing) and a single value of Q_0 for all three bands. The three parameter fit gave $Q_0 \approx 12.0$ eb,
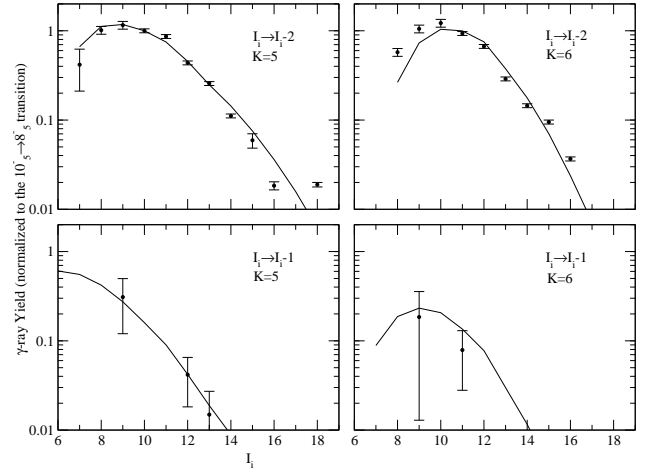
typical for nuclei in this mass region [26] and similar to previous measurements for ^{241,242,243}Am [28, 29].

IV. DISCUSSION

The proton-neutron configurations built on the proton $5/2^{-}[523]$ Nilsson orbital [22, 24] leading to the rotational bands in ^{242}Am are displayed in Figure 7, including the states identified in the present work. The $K^{\pi} = 5^{-} \rightarrow K^{\pi} = 6^{-}$ and $K^{\pi} = 5^{-} \rightarrow K^{\pi} = 3^{-}$ transitions are both K-allowed and involve a one-neutron excitation. They
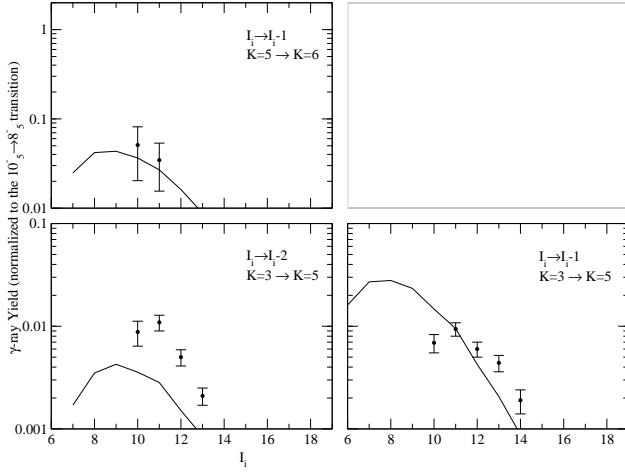
FIG. 6: Measured (points) and calculated (lines) interband γ-ray yields of the $K^\pi = 5^-$ $K^\pi = 3^-$ bands from the model-dependent fit of matrix elements described in the text.
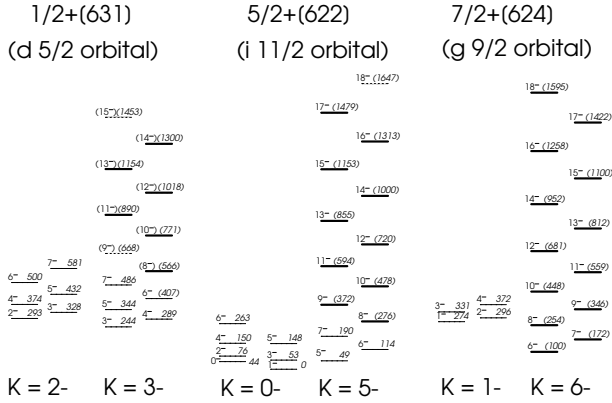

FIG. 7: The known multiplets resulting from coupling of the proton $5/2^-[523]$ Nilsson orbital (h 9/2) to each of three neutron orbitals, labeled with their Nilsson numbers and spherical shells. Previously known states (thin lines) are from [22]. Energies of new levels from the present work (bold lines) have errors of ≤ 2 keV.

would therefore be expected to have intrinsic matrix elements of similar magnitude, apart from the influence of K-mixing effects. However, the fit found interband intrinsic matrix elements $\langle K^\pi = 6^- \mid E2 \mid K^\pi = 5^- \rangle = 2.0$ eb and $\langle K^\pi = 3^- \mid E2 \mid K^\pi = 5^- \rangle = 0.5$ eb.

It might be expected that the nearly degenerate $K^\pi = 5^-$ and 6^- bands should be mixed by a first order ($\Delta K = \pm 1$) Coriolis effect. Coriolis interaction matrix elements derived from the $K^\pi = 5^-, 6^-$ level energies (Figure 3) and the calculations of Chasman et $al.$ over-predict the level splitting between the equal-spin members of the 5^- and 6^- bands by a factor of ≈ 3, a typical discrepancy for nuclei in the region [30]. However, the remarkably similar population strengths of the two bands imply strong $\Delta K = \pm 1$ mixing. A weaker Coriolis mixing of the $K^\pi = 2^-$ and 3^- bands would be expected,

since the energy splittings of the states are larger.

Mixing between the $K^\pi = 5^-$ and 3^- bands by Coriolis alignment would be a second-order ($\Delta K = 2$) effect [8], and this is consistent with the smaller $\langle K^\pi = 3^- \mid E2 \mid K^\pi = 5^- \rangle$ value from the fit. In addition, first order $\Delta K = 1$ mixing of a $K^\pi = 2^-$ component in the $K^\pi = 3^-$ band would be expected to lower the matrix elements to the 5^- band further. Salicio et $al.$, have measured transitions from the $I^\pi K = 3^-3$ and 4^-3 states to the $K^\pi = 0^-$ GSB and to the $K^\pi = 5^-$ isomer with similar γ-ray intensities. The $1 \leq \nu \leq 2$ K-forbidden $I^\pi K = 3^-3$ to $K^\pi = 0^-$ transitions have 15% to 50% γ-ray branches, compared to the 20% $I^\pi K = 3^-3 \rightarrow 5^-5$ branch, calculated from the measured γ-ray intensities and E2/M1 mixing ratios of [22]. The competitive strength of the $K^\pi = 3^-$ to $K^\pi = 0^-$ transitions is consistent with a $K^\pi = 2^-$ admixture in the $K^\pi = 3^-$ band (Figure 7).

The EM decay of the ^{242m}Am isomer is known to proceed by a highly hindered ($f_\nu \approx 1.6 \times 10^5$), once K-forbidden ($\nu = 1$) E4 transition [1] to the $K = 0$, $I^\pi = 1^-$ ground state. The large hindrance corresponds to a $B(E4)$ value of $\approx 10^{-5}$ single-particle units and reflects the purity of the $K = 5$ isomer and the $K = 0$ ground state. That is, small $K = 1$ admixtures in the $I^\pi K = 1^-0$ ground state or of $K < 5$ admixtures in the isomer state would result in a K-allowed term in the EM decay matrix element and a much shorter half life. Of course, the $I^\pi = 5^-$ isomer cannot mix with $K > 5$ configurations, and so it is expected to be a virtually pure $K = 5$ state.

Work is underway to reproduce the full set of branching ratio data which were not included in the first iteration of fits. A two-state mixing model is being evaluated as an alternative to the Alaga rule coupling for the $K^\pi = 5^-, 6^-$ bands and is expected to elucidate the strength of mixing between these two bands. This model is expected to improve the fit to the measured $K^\pi = 3^-$ band yields (Figure 6) as well. While the lowest states of the 3^- band populated in the present experiment are known to feed the ground-state band, direct evidence of Coulomb de-excitation to the ground state has yet to be found. The high level density of this odd-odd nucleus and the strong population of the $K^\pi = 5^-, 6^-$ states make observation of weak transitions difficult, and a systematic search for weakly-populated unknown states above the $I^\pi = 6^-$ state of the GSB is in progress. Knowledge of the strength of EM matrix elements which connect the $K^\pi = 5^-$ band to the GSB would provide a measure of the spin-dependence of K-mixing and would be beneficial in evaluating the possibility of depopulating the isomer by photon absorption or Coulomb excitation.

V. CONCLUSION

Coulomb excitation of a nearly pure ^{242m}Am isomer target has provided a significant increase in sensitivity to the states coupled to the isomer band. This has revealed

strong $\Delta K = 1$ mixing between newly discovered states up to $I \approx 18$ built on the previously known $K^\pi = 5^-$, $t_{1/2} = 141$ y isomer and a previously unidentified *yrast* $K^\pi = 6^-$ rotational band. The smaller $\Delta K = 2$ transition probabilities between the $K^\pi = 3^-$ and 5^- bands are consistent with Coriolis alignment, but the Coriolis interaction strength overpredicts the perturbations on the level energies by a factor of ≈ 3. While the higher-order spin-dependent $K \gg 1$ mixing phenomena observed in the Coulomb excitation of ^{178}Hf have not been detected in ^{242}Am, the much stronger $\Delta K = 1$ mixing in the present experiment would be expected to obscure the former effect. Previously unknown states assigned to a known $K^\pi = 3^-$ band were populated from the $K^\pi = 5^-$ isomer band at the $\sim 1\%$ level. This $K^\pi = 3^-$ band is known to feed the $I^\pi K = 1^-0$, $t_{1/2} = 16$ h ground state by γ decay at a similar strength to its feeding of the 5^- isomer, so the present Coulomb excitation implies the depopulation from an isomer state to a ground state. Previous measurements of $K^\pi = 3^-$ to $K^\pi = 5^-$ (K-allowed) and $K^\pi = 3^-$ to $K^\pi = 0^-$ (K-forbidden) γ-ray feeding of similar intensities suggest another instance of $\Delta K = 1$ mixing between the known $K^\pi = 2^-$ and 3^-

bands. A search for evidence of direct EM coupling of the upper $K^\pi = 3^-, 5^-$ states to the ground-state band is the subject of continuing investigation. An iterative fit of the matrix elements coupling the $K^\pi = 3^-, 5^-$ and 6^- bands and a comparison to the various theoretical descriptions are in progress to determine the strength and origin of observed K-mixing.

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